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RESEARCH ARTICLE

Geo-environmental assessment of geochemistry of groundwater and associated human health risks in the dry zone terrain of Sri Lanka

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Abstract: The close association between the geographic distribution of certain diseases and environmental factors is clearly apparent in the tropical terrain of Sri Lanka. Among these diseases, dental and skeletal fluorosis and Chronic Kidney Disease of uncertain aetiology (CKDu) are common problems in the dry zone regions of the country. The quality of drinking water is primarily blamed for the emergence of such diseases. Since 2015, about 1250 groundwater samples from the dry zone region of Sri Lanka were collected and analyzed for their water quality parameters. Excessive ionic concentrations were observed in groundwater in which solute composition is dominated by Na^+, K^+, Ca^{2+}, Mg^{2+}, HCO_3^-, Cl^-, and SO_4^{2-}. The results revealed that the dry zone was highly affected by excessive levels of fluoride and divalent cations. Fluoride content in the study samples varied up to 6.8 mg/L. About 35% and 73% of samples from metamorphic and sedimentary terrains, respectively, were identified as very hard and not palatable. Higher As content is a significant feature in groundwater from the sedimentary terrain in the north and northwest of the island. Excessive nitrate was also noted in highly permeable sandy aquifers in the coastal belt. Results of available water quality data confirm that the groundwater quality of the dry zone is affected by both geogenic and anthropogenic factors, while certain geographically distributed health issues are closely associated with the groundwater geochemistry.

Keywords: Arsenic, CKDu, dental fluorosis, excess fluoride, medical geology, water hardness.

INTRODUCTION

For centuries, it has been known that human health is strongly influenced by the natural environment, in which geological factors play a crucial role in determining the well-being of biota. In this view, the emerging interdisciplinary science of 'medical geology' addresses the interaction between natural geological factors and associated health effects on humans and animals (Dissanayake & Chandrajith, 1999; Bunnell, 2004; Dissanayake, 2005). The availability of trace elements and their species in the geological environment varies widely. The availability of such components may have a beneficial impact on human health, but excessive content leads to detrimental health effects. For instance, it is well-known that human health can be adversely affected by exposure to contaminants such as As, Cd, Hg, Pb, and fluoride. Natural geogenic processes and anthropogenic activities contribute contaminants to the environment at different levels (Bundschuh *et al*., 2017). Biotic and abiotic interactions that occur in the geosphere leads to the accumulation and magnification of elements and their species in trophic chains and food webs, resulting in either beneficial or detrimental effects on organisms (Dissanayake, 2005).

Drinking water is perhaps the most vital natural resource, and an estimated half of the world's population, particularly in developing countries entirely rely on groundwater for drinking (Kløve *et al*., 2014). Over 66% of households in rural Asia-Pacific regions use groundwater for drinking (Carrard *et al*., 2019). Not only that, groundwater is extensively extracted in this region for irrigation purposes. For instance, 38% of agricultural lands in the world are currently irrigated with groundwater

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(Siebert *et al*., 2010). For all these purposes, the quality of water is extremely vital and the requirement of clean water is one of the fundamental commodities for human life. Hence, monitoring groundwater quality is one of the most important aspects of water resources management and supply of suitable quality water for human consumption. Particularly in developing countries, the majority of the population depends on groundwater that is extracted and consumed without any pre-treatment. Clean water is often scarce in such regions, and hence groundwater quality monitoring is extremely important. As set forth by the United Nations, access to clean water and sanitation (SDG 6) and good health and well-being (SDG 3) are important aspects of the Sustainable Development Goals (SDGs) that need to be achieved by the year 2030 (UN 2015).

There are ample reports on geographically distributed health issues and their close association with the geo-environment, especially relevant to drinking water quality. For instance, high arsenic (As) and fluoride (F^-) in groundwater have been major public health concerns for several decades (Karim, 2000; He *et al*., 2013; Rodríguez-Lado *et al*., 2013; Wen *et al*., 2013). Such health issues are more apparent in countries in the equatorial belt, which are having unique geological and climatological characteristics and are mostly considered as less developed with many socio-economic issues. These tropical regions continuously experience intense rainfalls and prolonged droughts, which facilitate geochemical partitioning, resulting in depletion or enrichment of elements (Dissanayake & Chandrajith, 1999). This might be more severe in the near future due to predicted climate change impacts, and more effects may focus on water quality and water stress in many regions (Kundzewicz *et al*., 2008).

Sri Lanka is an Indian Ocean island with a population of 22 million, of which only about 44% have access to a pipe-borne water supply, while the rest mainly depend on the tube and dug wells (Fan, 2015). With its varied geology, topography, and climate, the quality of water also varies widely. Although the island is small, with only about 65,000 sq km land area, distinct rainfall regimes with different climatic conditions can be identified. From which, the wet zone receives an average annual rainfall of about 2500 mm, while the dry zone receives significantly lower annual precipitation (<1500 mm) (Figure 1). Over 60% of the land area of Sri Lanka belongs to the dry zone where rainfall is restricted to a few months of the year. In this climatic region, most households depend on groundwater for drinking. In some regions, the entire population depends on groundwater for their daily needs. As a country in the tropical belt, a close relationship between human health and the immediate natural environment is more visible in Sri Lanka (Dissanayake & Chandrajith, 1999). In the last few decades, several studies were carried out in Sri Lanka to illustrate the impact of natural geological factors and the distribution of certain chronic diseases, health, and nutrition. Groundwater fluoride and dental and skeletal fluorosis (Dissanayake, 2005; Chandrajith *et al*., 2012; 2020), nitrate in drinking water and related health impacts (Dissanayake & Weerasooriya, 1987), iodine and iodine-deficient disorders (IDD) (Dissanayake & Chandrajith, 1993), and water hardness and occurrence of urinary calculi (Abeywickarama *et al*., 2016) are among many others. Over the last two decades, a new form of Chronic Kidney Disease without any known etiological factors (CKDu) has emerged in the dry zone of Sri Lanka and is also considered to be associated with the quality of drinking water (Chandrajith *et al*., 2011b; Wickramarathna *et al*., 2017; Nanayakkara *et al*., 2019; Balasooriya *et al*., 2020; Liyanage *et al*., 2022). These facts suggest Sri Lanka as an ideal location to investigate the geochemistry of groundwater, which is a very useful tool for delineating regional health issues and identify the geographic distribution of chronic diseases. The aetiology of certain geoenvironment-related diseases can be delineated effectively with the help of an investigation of the geochemical composition of groundwater. Therefore, the need of monitoring the groundwater quality in Sri Lanka is of foremost priority in achieving SDGs by the year 2030. In the dry zone regions of Sri Lanka, over 87% of the community depends on groundwater extracted from shallow dug wells (<10 m) and deep tube wells for their domestic needs (Dissanayake & Chandrajith, 2018). The utilization of groundwater for drinking with no pre-treatment indicates a link between groundwater geochemistry and certain health issues. Therefore, this paper aims to present hydrogeochemical data giving particular attention to the dry zone area where most of the environment-related health issues are widely reported. The improved understanding of the quality of groundwater is important in identifying possible relationships with geographically distributed health issues, and also for water resource management for ensuring the sustainable use of groundwater to achieve the proposed SDGs.

Climate, geology, and groundwater

Sri Lanka has a humid tropical climate with monsoon rainfall and well-demarcated spatial variation. The southwest monsoon brings heavy rains into the southwest wet zone region with over 2500 mm/a precipitation while the northeast monsoon mainly falls in the dry zone with about 1000 mm/a rain. In between these two climatic zones, an intermediate zone is also characterized with 1700 mm/a precipitation while in the dry zone, evaporation exceeds the precipitation (Jayasena *et al*., 2008). Geologically, over 90% of the island of Sri Lanka comprises amphibolite and granulite facies metamorphic rocks, and the groundwater in this terrain is mostly extracted through deep (tube) and shallow wells (Dissanayake & Chandrajith, 2018). Shallow wells are often constructed in the unconsolidated overburden, while deep wells penetrate to fractures, cracks, and fissures in the hard rocks. A marked difference in physicochemical parameters can also be observed between shallow and deep wells, although a seasonal variation of water quality is not apparent (Wickramarathna *et al*., 2017; Senarathne *et al*., 2021). A narrow terrain in the northern and north-western coastal belt is underlain by Miocene limestone sequences that overlay Quaternary deposits, mainly dune sands. In this region, groundwater is mostly extracted by shallow wells penetrated to either karst aquifers or groundwater trapped in perch water lenses in dune sand. The water quality in these sedimentary aquifers is drastically different from groundwater from the metamorphic terrain (Thilakerathne *et al*., 2015).

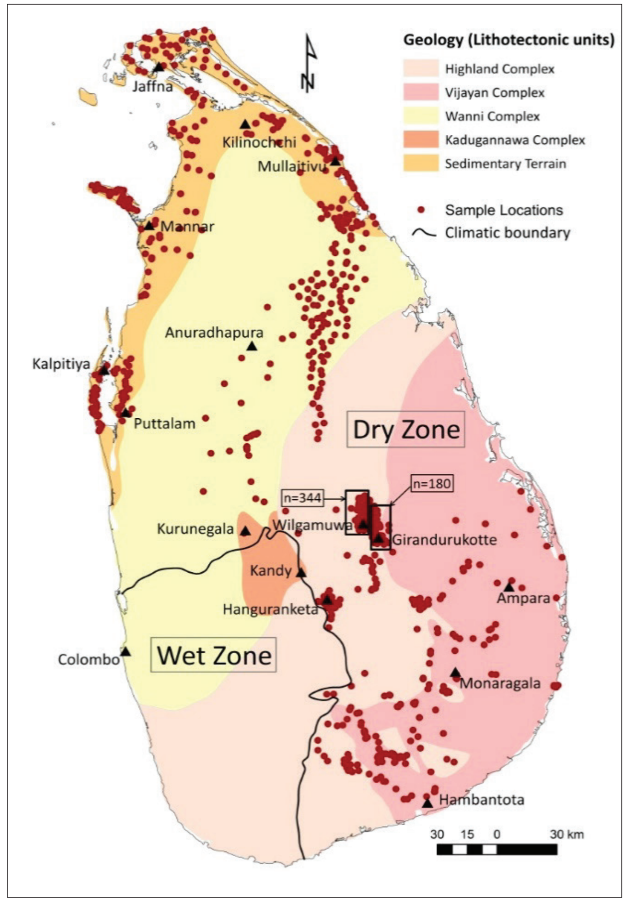

Figure 1: Geological, climatological characteristics of Sri Lanka and sampling locations

METHODOLOGY

Sample collection

Since 2015, extensive investigations on groundwater geochemistry in the dry zone terrain of Sri Lanka have been carried out by the Department of Geology, University of Peradeniya, Sri Lanka. During this period over 1250 groundwater samples were collected which 966 samples were obtained from regolith aquifers in the metamorphic terrain (Figure 1). In this study, primary and secondary data were considered together and analyzed to obtain a comprehensive overview of the water quality in the dry zone of Sri Lanka. Secondary data for this work were obtained from previous studies (Chandrajith *et al*., 2016; Wickramarathna *et al*., 2017; Bandara *et al*., 2018; 2020; Jayathunga *et al*., 2020; Udeshani *et al*., 2020; Senarathne *et al*., 2021; Liyanage *et al*., 2022).

Analytical methods

For the analyses of all water samples, methods described by the American Public Health Association (APHA, 1998) were used. The pH and Electrical Conductivity (EC) were measured using field-portable EC-pH meters. Filtered and unacidified samples were used for anion analysis. For cation analysis, samples were filtered using 0.45 µm nylon filters and subsequently acidified with 1% v/v HNO_3. All samples were kept in the dark at 4 °C until analyses were performed. Alkalinity and hardness were determined by sulphuric acid and EDTA titrimetric methods, respectively. Anions, mainly fluoride, chloride, nitrate, and sulphate contents were measured either by ion chromatography (Thermo Dionex ICS-1100) or using a visible spectrophotometer (Hach 2400). Major cations (Na^+, K^+, Ca^{2+}, Mg^{2+}) were measured by atomic absorption spectrometry (Varian 240FS). In addition, trace metals in groundwater samples were measured by Inductively Coupled Plasma Mass Spectrometry (Thermo Scientific iCAPQ). Detailed procedure and quality control adopted in sample analysis were described in Balasooriya *et al*. (2020). Concentrations below the detection limits were allocated a value of half of the limit of detection (LOD) during statistical interpretations. The analytical precision was determined by calculating the cation-anion balance considering major cations (Na^+, K^+, Ca^{2+}, and Mg^{2+}) and anions (F^-, Cl^-, NO_3^-, HCO_3^-, and SO_4^{2-}) and it was within 10% in most cases.

RESULTS AND DISCUSSION

Geochemical characteristics of dry zone groundwater

The summary results of the analyzed physicochemical parameters are given in Table 1. The pH of the groundwater in the dry zone region of Sri Lanka varied from slightly acidic to alkaline. In the metamorphic terrain, the median pH was 6.75 (± 0.65); however, a higher median pH (7.42 ± 0.66) was observed in groundwater in the sedimentary terrain. Higher evaporation rates and dissolution of minerals in soils and rocks may cause increased pH in groundwater. The electrical conductivity (EC) varied from 4.94 µS/cm to 5.22 mS/cm in the metamorphic terrain, with a median of 324 µS/cm, implying a higher rate of mineralization. According to Sri Lanka Standards (SLS), an EC of 750 µS/cm is considered as the maximum permissible limit for drinking water (SLS, 2020); 21% of the dry zone samples from the metamorphic terrain exceeded this level. Further, 39% and 20% of samples were categorized into medium salinity (250–750 µS/cm) and high salinity (750–2250 µS/cm) groups, respectively, based on the EC classification of Handa (1969). Interestingly, comparatively higher EC values were reported in the sedimentary terrain that varied from 16.2 µS/cm to 10.9 mS/cm with a median of 1056 µS/cm, while 63% of the samples exceeded the maximum permissible limit for drinking water (Figure 2). Higher amounts of dissolved solids in groundwater from the sedimentary terrain resulted from the dissolution of carbonate rocks and the mixing of saline water (Chandrajith *et al*., 2016; Bandara *et al*., 2018).

The major solute composition of all the dry zone water is dominated by Na^+, K^+, Ca^{2+}, Mg^{2+}, HCO_3^-, Cl^-, and SO_4^{2-}. In most wells, anion abundances followed the sequence $HCO_3^- > Cl^- > SO_4^{2-} > NO_3^-$. Chloride contents in the metamorphic terrain varied from 0.20 to 3100 mg/L with a median of 25.0 mg/L. However, only 3.6% of the samples collected near the coastline exceeded the recommended limit. In contrast, 45% of the samples from the sedimentary terrain showed higher Cl^- levels that are mainly due to seawater intrusions. The median sulphate of groundwater in the metamorphic terrain was 12.2 mg/L, while it was 37.4 mg/L in the sedimentary terrain. Accordingly, 14% of the samples from sedimentary terrain showed higher sulphate contents (>250 mg/L), and exceptionally higher values (>500 mg/L) were mostly observed in the Kalpitiya region. This is mainly due to the extensive application of chemical fertilizer in the region (Jayathunga *et al*., 2020).

Among the investigated anionic constituents, a comparatively higher nitrate content was noted in the dry zone sedimentary terrain at a median of 1.20 mg/L, while it was 0.90 mg/L in the metamorphic terrain. The primary sources of nitrate in groundwater are fertilizer applications, septic tank leachates, and leaching from open waste dumps. Although agriculture is widespread throughout the dry zone terrain, all the values reported in the crystalline rock terrain were well below the recommended limits (50 mg/L as NO_3^-). In contrast, aquifer materials of sedimentary terrain are highly permeable and can easily be contaminated due to anthropogenic activities. This is clear in the reported highest nitrate values, which exceed the maximum permissible level in the Kalpitiya region where extensive fertilizer applications are reported. In addition, as evident from water isotope signatures, irrigation return flows can also increase the nitrate content with the continuous applications of nitrogenous fertilizers in areas that are underlain by sedimentary rocks and sediments (Chandrajith *et al*., 2016; Jayathunga *et al*., 2020).

The hardness of water is one of the main characteristic features of the groundwater in the dry zone terrain of Sri Lanka. The median hardness (as $CaCO_3$ equivalent) reported in the metamorphic terrain was 130 mg/L whereas 286 mg/L was reported in the sedimentary terrain (Table 1). From among samples from the metamorphic terrain, 28% and 19% fell in the moderately hard (61–120 mg/L) and hard (121–180 mg/L) categories, respectively. Nearly 35% of the samples can be categorized as very hard water (>180 mg/L) (Figure 2 and 3). The recommended range for hardness in drinking water is between 100 and 150 mg/L and the maximum desirable limit is 250 mg/L (SLS, 2020). Among all collected samples, 23% exceeded the 250 mg/L limit while in the sedimentary terrain, 73% of the samples were very hard and 59% exceeded the 250 mg/L limit.

Table 1: Major element composition of groundwater in studied wells

Parameter	Metamorphic terrain							Sedimentary terrain							Maximum permissible level	
	Mean	Median	SD	Min	Max	Percentile 25	Percentile 75	Mean	Median	SD	Min	Max	Percentile 25	Percentile 75	Sri Lankan Standards; SLS 614, 2013	WHO Standards; WHO, 2022
pH	6.74	6.75	0.65	4.49	8.96	6.27	7.18	7.37	7.42	0.66	5.10	9.36	7.10	7.79	6.0 - 8.5	6.5 - 8.5
EC (µS/cm)	492	324	526	4.94	5224	154	666	1720	1056	2020	16.2	10900	462	2145	750	1500
TDS (mg/L)	504	344	533	12.9	5442	156	712	1293	792	1284	69.8	5950	495	1592		
Alkalinity (mg/L)	214	150	203	3.60	1865	76.0	324	214	214	171	1.60	826	39.2	340	200	500
HD (mg/L)	180	130	176	4.00	2210	72.9	234	475	286	634	16.9	5810	157	518	250	300
Anions (mg/L)																
NO_3^-	1.78	0.90	2.63	< 0.02	27.0	0.50	2.20	12.4	1.20	63.4	< 0.02	845	0.50	4.85	50	50
SO_4^{2-}	22.8	12.2	42.9	< 0.05	690	6.00	24.4	126	37.4	254	0.05	2166	14.8	120	250	250
PO_4^{3-}	0.33	0.21	0.58	< 0.01	9.90	0.06	0.37	0.88	0.44	1.58	< 0.01	11.3	0.26	0.81	2	0.1
Cl^-	57.1	25.0	149	< 0.20	3100	13.9	49.8	464	190	708	5.55	4830	64.1	561	250	250
F^-	0.82	0.57	0.83	< 0.02	6.80	0.29	1.08	0.51	0.39	0.52	< 0.02	5.04	0.17	0.68	1	1.5
Major elements (mg/L)																
Na	43.6	24.4	62.9	0.08	654	11.4	46.9	148	61.2	249	6.37	1700	34.4	130	250	200
K	2.83	1.39	4.55	0.12	51.0	0.72	2.90	14.9	6.91	25.1	0.27	267	2.84	18.0	ND	20
Ca	62.1	32.1	113	0.20	1451	16.1	61.8	117	76.9	151	2.96	1164	35.8	141	100	75
Mg	19.3	12.5	26.0	0.05	472	5.65	24.4	33.5	20.0	47.8	1.53	470	9.30	36.5	30	50
Trace elements (µg/L)																
Li	3.27	1.43	5.59	< 0.02	50.3	0.71	3.22	12.6	4.13	20.1	< 0.02	147	1.27	15.8	ND	ND
Al	44.9	9.03	176	< 0.05	2561	4.01	20.1	38.1	9.20	136	0.16	1680	4.71	23.6	200	200
Cr	1.78	0.38	6.30	< 0.01	88.1	0.16	1.04	7.13	0.52	43.2	0.01	599	0.23	1.14	50	50
Mn	145	33.9	337	0.01	4542	6.87	119	126	41.6	254	0.45	2268	7.31	129	100	80
Fe	252	25.1	783	< 0.01	8152	8.72	82.8	463	52.4	3082	0.82	46197	19.4	118	300	300
Co	0.77	0.15	1.91	0.01	17.2	0.06	0.52	1.43	0.27	4.9	0.02	67.7	0.10	1.02	ND	ND
Ni	26.4	1.74	112	0.01	1129	0.90	3.80	7.61	2.26	17.3	0.19	145	1.24	6.57	20	70
Cu	8.54	0.80	36.1	0.01	432	0.40	1.95	9.05	1.50	45.2	0.01	560	0.77	2.96	1000	2000
Zn	115	24.9	1113	0.03	32176	9.75	53.9	88.8	26.4	206	0.35	2332	12.5	79.2	3000	3000
As	0.35	0.20	0.53	< 0.02	8.14	0.10	0.39	3.07	1.01	5.6	0.03	46.9	0.34	3.39	10	10
Se	0.66	0.27	1.27	< 0.02	14.6	0.07	0.69	1.82	0.57	2.8	< 0.02	20.8	0.25	2.11	10	40

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Parameter	Metamorphic terrain							Sedimentary terrain							Maximum permissible level	
	Mean	Median	SD	Min	Max	Percentile		Mean	Median	SD	Min	Max	Percentile		Sri Lankan Standards; SLS 614, 2013	WHO Standards; WHO, 2022
						25	75						25	75		
Rb	4.58	1.21	15.9	0.16	220	0.65	3.10	11.8	9.75	12.1	0.34	76.1	3.19	16.8	ND	ND
Sr	353	226	403	4.30	4016	118	443	1339	765	2048	28.4	21000	326	1459	ND	ND
Cd	0.29	0.03	1.96	< 0.01	43.0	0.01	0.09	0.11	0.07	0.2	< 0.01	3.44	0.03	0.10	3	3
Ba	140	83.1	161	1.40	1477	46.5	169	146	96.4	153	2.90	1124	42.1	190	ND	700
Pb	0.83	0.15	3.51	< 0.01	65.4	0.06	0.42	0.94	0.12	2.9	< 0.01	37.2	0.07	0.79	10	10

EC- electrical conductivity; TDS- total dissolved solids; HD- total hardness; ND- not defined

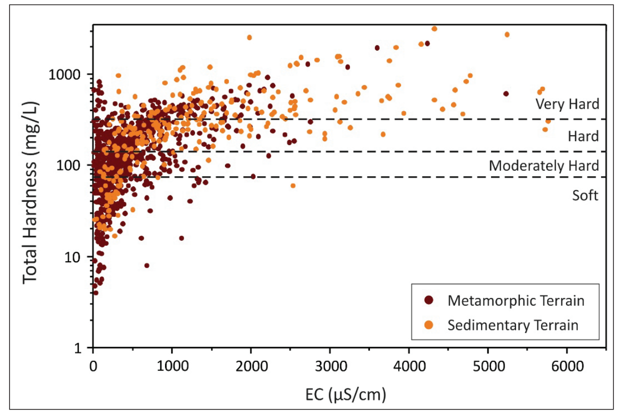

Figure 2: Relationship between water hardness and electrical conductivity (EC) of groundwater in the study area

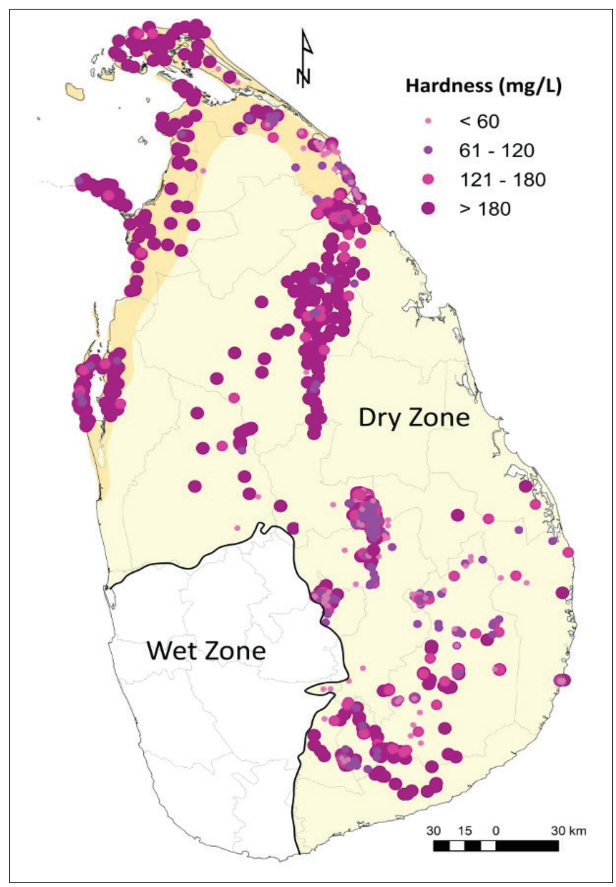

Figure 3: Distribution of the groundwater hardness in the dry zone of Sri Lanka

Fluoride is an important hydrogeochemical parameter that enters the human body primarily from drinking water, and hence the geochemistry of fluoride in a region is of extreme importance. The dry zone of Sri Lanka is well-known for a higher content of fluoride in drinking water, with reported health issues of dental and skeletal fluorosis (Dissanayake & Chandrajith, 1999; Dissanayake, 2005; Ranasinghe *et al*., 2019b). The fluoride content in hard rock terrain varied from < 0.01 to 6.80 mg/L with a median of 0.57 mg/L, of which 35% exceeded the recommended value of 0.8 mg/L for tropical countries (WHO, 2004) while 15% of those samples exceeded the 1.5 mg/L limit (WHO, 2011). In the sedimentary terrain, the fluoride content varied from < 0.01 to 5.04 mg/L with a median of 0.39 mg/L, of which 17% exceeded the 0.8 mg/L limit (Figure 4). However, Warnakulasuriya *et al*., (1992) inferred that dental fluorosis is common among school children who drink water over 0.8 mg/L of fluorides.

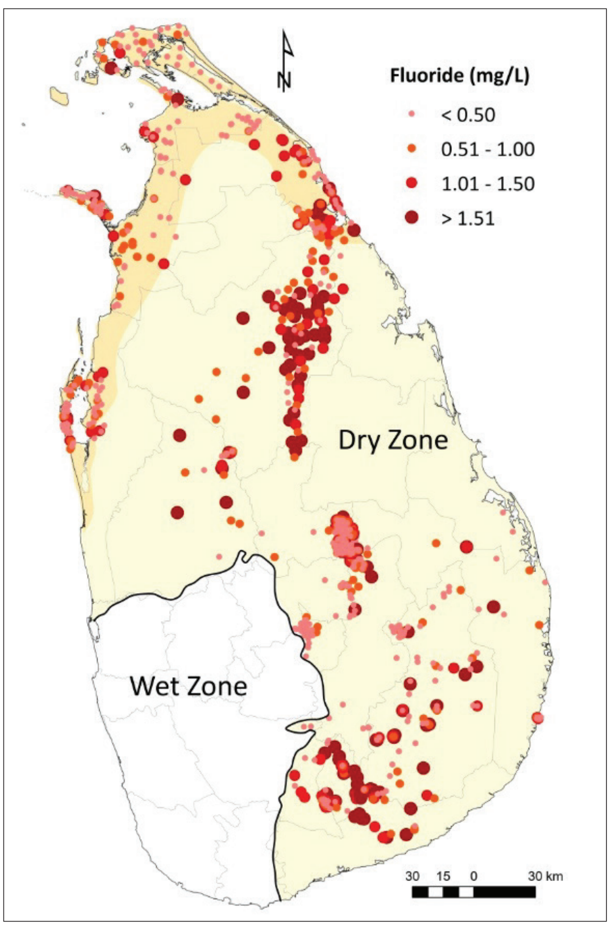

Figure 4: Distribution of the groundwater fluoride in the dry zone of Sri Lanka

The geochemical distribution pattern of fluoride in Sri Lanka depends on the source of groundwater, mineralogical composition of rocks and soils, and the climatic pattern (Dissanayake & Chandrajith, 1999; Chandrajith *et al*., 2012). In contrast, groundwater in the wet zone showed remarkably lower fluoride content. Higher rainfall in the wet zone results in rapid leaching of fluoride ions from rocks and soils; the fluoride is then washed away (Dissanayake, 1996). The rocks in the metamorphic terrain comprise fluoride-bearing minerals, mainly apatite, micas, and amphibolites, and the ion exchange with hydroxyl groups (OH^-) eventually increases the fluoride (F^-) leaching into groundwater. Higher pH, the presence of bicarbonates, and the availability of alkali and alkaline earth elements also contribute to the increased mobility of fluoride into groundwater (Saxena & Ahmed, 2003; Jacks *et al*., 2005). Additionally, higher evaporation under a tropical climate may enrich the fluoride content in groundwater (Dissanayake, 1996; Mukherjee & Singh, 2020). Although fluoride-bearing minerals are absent in the terrain underlain by the Recent deposits and Miocene limestone sequences, the application of phosphate-bearing fertilizer in these regions may increase the fluoride content in groundwater. Phosphate minerals are a well-known host for fluoride (Edmunds & Smedley, 2013).

Among the major cations, Na^+, Ca^{2+}, and Mg^{2+} are dominant in groundwater from the dry zone region. In the metamorphic terrain, the Na^+/Cl^- molar ratio was found to be >1, which implies the release of Na^+ during the process of silicate weathering (Figure 5). The ion exchange of Na^+ and K^+ in rocks with Ca^{2+} and Mg^{2+} in water is evident in this process (Li *et al*., 2008). The presence of Ca^{2+} and Mg^{2+} ions that contribute to the water hardness are considered to be an important parameter as it may possibly influence health issues such as the occurrence of urinary calculi (Abeywickarama *et al*., 2016). In the metamorphic terrain, 11% and 18% of the samples exceeded the maximum permissible levels for Ca^{2+} and Mg^{2+}, respectively. In the case of limestone terrain, 38% of the samples contained higher Ca^{2+} levels, exceeding the 100 mg/L limit, while 33% showed Mg^{2+} exceeding the 30 mg/L limit. Significant correlation of Ca^{2+} and Mg^{2+} with HCO_3^- signifies the dissolution of Ca-Mg bearing minerals in the metamorphic terrain. The interrelationship of ion ratios and salinity of groundwater in the Gibbs diagram (Gibbs, 1970) showed that the geochemistry of most of the dry zone water is governed by rock water interaction and silicate weathering and carbonate dissolution as key processes (Figure 6).

Weathering of the underlying rock and associated ion exchange processes are the key processes behind the major ionic constituents in groundwater. Some of the dry zone water, particularly from the sedimentary terrain, is influenced by evaporation and mixing of saline water, in agreement with the locations and prevailing dry climate.

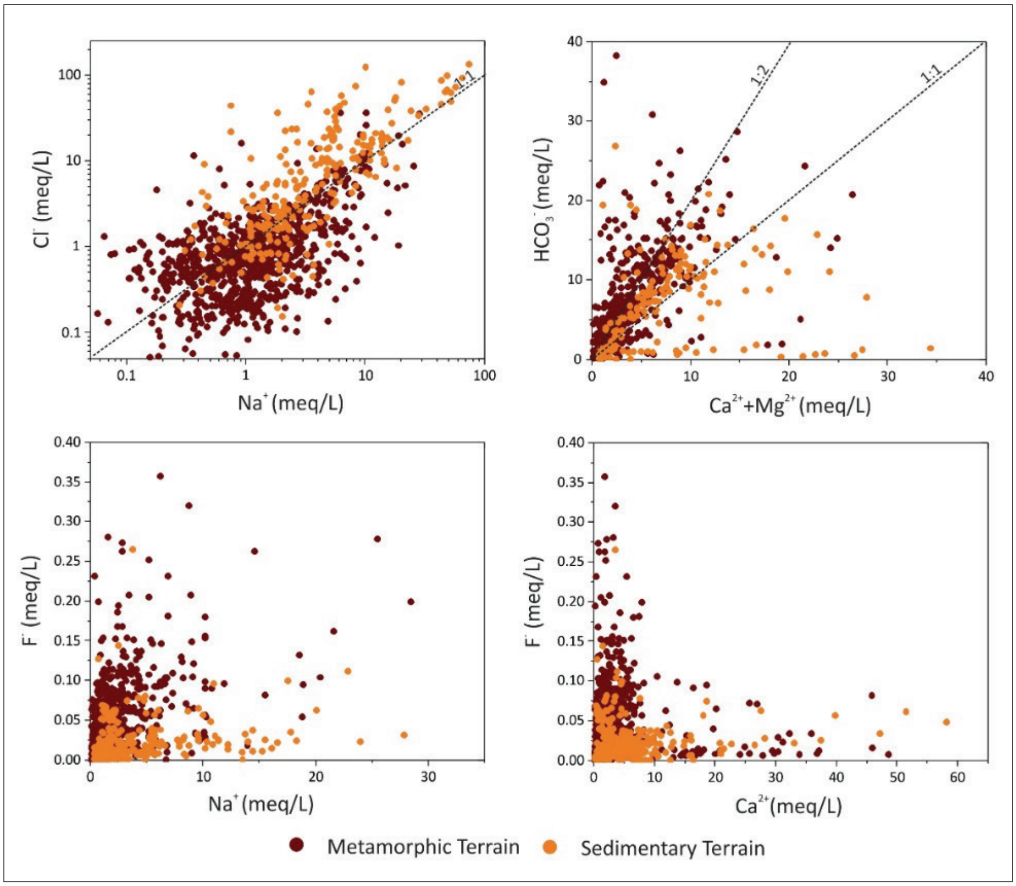

Figure 5: Ion scatter plots of major parameters in groundwater

Trace elements in groundwater

From a health perspective, some of the trace elements can cause toxic impacts on humans and animals even at very low concentrations. This situation is more problematic in areas where the community relies mostly on groundwater for their drinking purposes. The content of most toxic trace elements in groundwater was lower than the Sri Lankan drinking water quality standards and also the standards prescribed by WHO (2011) and USEPA (2012). Among the studied trace elements, Sr and Ba contents dominated in groundwater compared to other trace elements in both geological terrains (Table 1). The median content of total iron (Fe) in groundwater from metamorphic and sedimentary terrains was 25.1 μg/L and 52.4 μg/L, respectively, and 14% of the studied wells from each terrain exceeded the desired levels of iron in drinking water (0.3 mg/L). From among the top ten trace elements of major public concern, arsenic, cadmium, and lead in groundwater have become a global concern due to their high toxicity, persistence, and bio-accumulative nature (WHO, 2021). However, none of the samples from the metamorphic terrain exceeded the permissible limit for arsenic (10 μg/L). However, 6% of the samples from the sedimentary terrain exceeded the 10 μg/L limit; the majority of these samples were collected from the Mannar island. In most samples, Cd levels were at or below the detection limit of the ICP-MS (0.01 μg/L).

The Pb content in the majority of samples from both metamorphic and sedimentary terrains was well below the permissible limit of 10 μg/L, however, 13 samples from the metamorphic terrain and 2 from the sedimentary terrain exceeded the 10 μg/L limit. Selenium (Se) which is also known as an essential trace nutrient, was found to vary from < 0.02 μg/L to 14.6 μg/L in the metamorphic terrain and 0.02 to 20.8 μg/L in sedimentary terrain with median values of 0.27 μg/L and 0.57 μg/L, respectively.

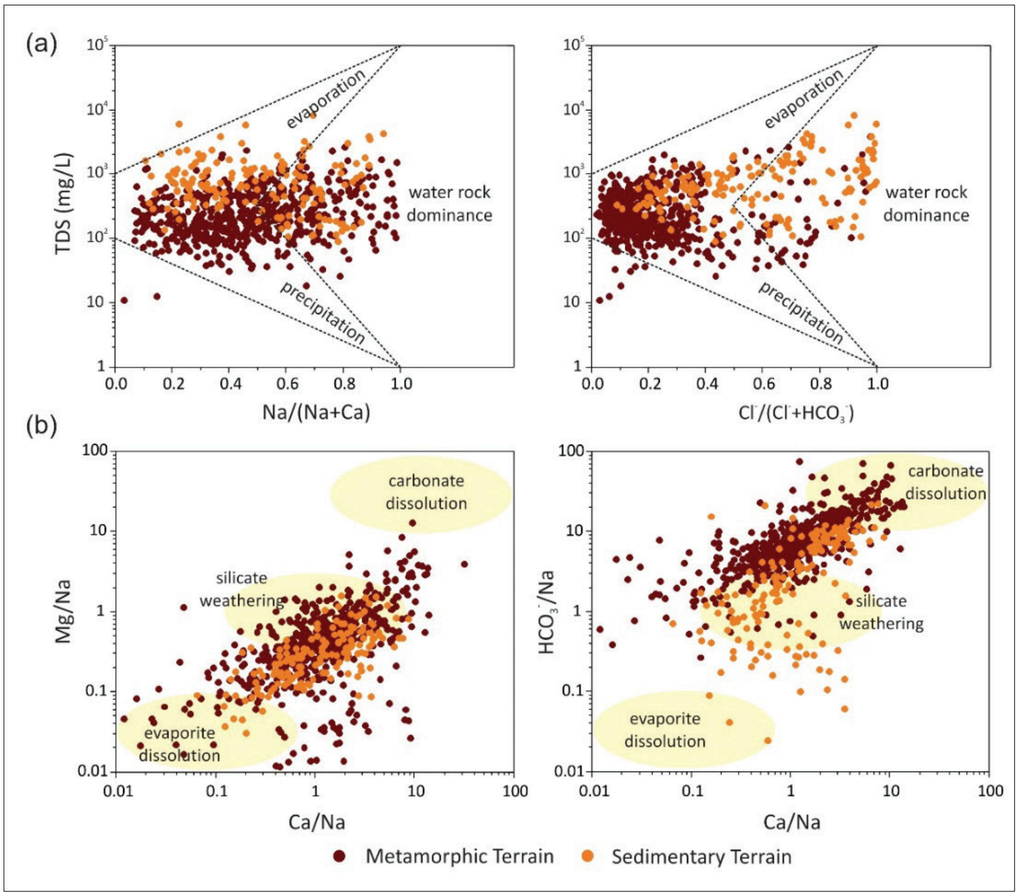

Figure 6: (a) Gibbs plot and (b) bivariate plot explaining the geochemical processes

Possible influence on health issues

Fluoride

It is well known that the lack of fluoride in drinking water causes tooth decay. On the other hand, high concentrations of fluoride can also lead to detrimental health issues. Dental fluorosis (mottling of teeth) and skeletal fluorosis (crippling of bones) can occur when the drinking water fluoride levels exceed 1.5 mg/L and 4.0 mg/L, respectively (WHO, 2011). However, these levels do not apply to tropical terrain where people consume more water due to local environmental conditions. For instance, health effects of fluoride have been observed even at low-level exposures particularly in tropical regions (Ghosh *et al*., 2013). Therefore, the limit of 0.8 mg/L has been defined for tropical climates by the WHO (2004). Although fluoride is beneficial in preventing dental caries by re-mineralization of enamel, it is not considered as an essential nutrient for human well-being, as it is not found in natural human metabolic pathways and no deficiency diseases have ever been reported (Peckham & Awofeso, 2014).

In the dry zone of Sri Lanka, excessive fluoride content in groundwater has been identified as a serious issue, as it compromises the oral health of children (Figure 7). Nearly 80% of the children from dry zone areas are affected by dental fluorosis (Van Der Hoek *et al*., 2003; Rajapakse *et al*., 2017). Moreover, some early studies provided evidence for severe forms of dental fluorosis (Ekanayake & van der Hoek, 2002). Ranasinghe *et al*. (2019b) revealed that 11.2% of the dry zone population (2.2 million) is at risk of potential health issues posed by high fluoride ingestion (>1.0 mg/L) via drinking water. The results of the present study also confirm the severity of the issue, as it revealed that 31% of the groundwater samples showed excess fluoride levels. Long-term consumption of such water may enhance the prevalence of dental fluorosis and also may cause an array of health issues. However, a heterogeneous distribution of fluoride content is depicted in the studied wells in which wells with low fluoride (< 0.5 mg/L) were located close to wells with higher fluoride (Figure 4). Ranasinghe *et al*. (2019b) identified that the minimum distance between a well with high fluoride (> 4.0 mg/L) and low fluoride (< 1.5 mg/L) is closer to 42 m. Since the effect of seasonal variation of fluoride in dry zone wells is negligible (Wickramarathna *et al*., 2017), the wells with safe fluoride can be used as sources of a drinking water supply to the inhabitant population.

Figure 7: A typical case of dental fluorosis in the dry zone of Sri Lanka

The emerging incidence of chronic kidney disease with undetermined origin (CKDu) is one of the main health issues that contribute significantly to high morbidity and mortality rates among dry-zone farming communities in Sri Lanka (Vlahos *et al*., 2019; Fernando *et al*., 2020a). Thousands of deaths have been reported in the dry zone of Sri Lanka during the past 2-3 decades due to CKDu and other kidney-related health problems (Ranasinghe *et al*., 2019a). The prevalence of CKDu between 15.1 and 22.9% has been observed among the residents in affected regions (Jayatilake *et al*., 2013; Kafle *et al*., 2019; Vlahos *et al*., 2019). The geographic distribution of disease shows an uneven patchy pattern in which CKDu villages are located adjacent to non-CKDu villages (Balasooriya *et al*., 2020). The inferior quality of the drinking water consumed by the affected community is the most suspected cause for the disease (Chandrajith *et al*., 2011b; Wickramarathna *et al*., 2017; Nanayakkara *et al*., 2019; Balasooriya *et al*., 2020; Liyanage *et al*., 2022). To a certain extent, the areas with CKDu overlap with the areas with high fluoride levels (Figure 4 and 8) and the excess fluoride intake

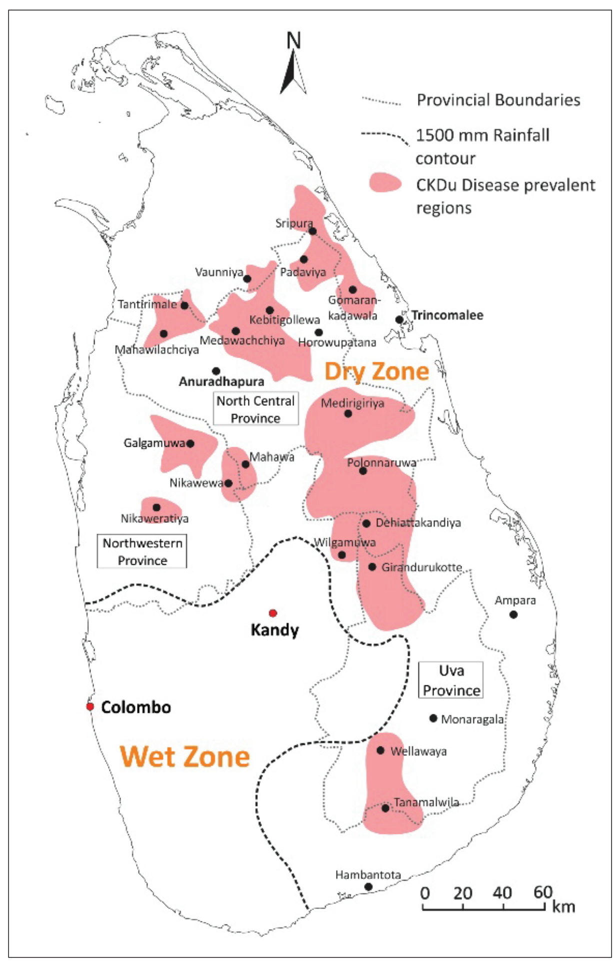

Figure 8: Identified CKDu hotspots of the dry zone of Sri Lanka

can lead to kidney damage (Dissanayake & Chandrajith, 2017; 2019). Fluoride excretion from the body makes kidneys a target organ for fluoride accumulation and the nephrotoxic nature of fluoride has been proven in previous studies (Chattopadhyay *et al*., 2011; Niu *et al*., 2016). It was also reported that serum fluoride levels are higher in CKDu patients compared to the control group in the same area (Fernando *et al*., 2020b). Molecular-level damage can occur in kidneys due to long-term exposure to fluoride, and impaired kidneys may retain more fluoride, which may enhance the damage (Perera *et al*., 2021). When considering the threshold value for fluoride defined for the dry zone of Sri Lanka (0.6 mg/L), 48% of the samples from metamorphic terrain showed higher concentrations, alone with a high hardness that is believed to behave synergistically to damage kidneys (Dissanayake & Chandrajith, 2019; Perera *et al*., 2021). Although the water hardness in the sedimentary terrain, where limestone is predominant, is higher, fluoride levels are much lower in many instances, while CKDu is not reported in these areas.

Nitrates

Although nitrate-related health issues are not investigated in depth in Sri Lanka, a significant correlation between nitrate concentration in groundwater and various types of human cancer such as stomach, small intestine, oesophagus, and liver cancers, total malignant incidence, and benign tumors was reported (Dissanayake & Weerasooriya, 1987). However, the levels of nitrate in groundwater are vitally important as most of the dry zone communities rely on groundwater as their drinking water source. Since rice and vegetable farming is widespread, extensive application of nitrogenous fertilizer can induce the levels of nitrate in the dry zone area. The nitrate derived from fertilizer is readily soluble and can easily leach from the soil. Particularly, the use of nitrogenous fertilizers such as urea $[CO(NH_2)_2]$, ammonium sulphate $[(NH_4)_2SO_4]$, and ammonium nitrate (NH_4NO_3) and mixing of groundwater with irrigated return flows can lead to extreme nitrate levels (Jayathunga *et al*., 2020). As depicted in the Kalpitiya region, the effect of fertilizer on higher nitrate levels may visible over a large area, with up to 846 mg/L nitrate-N in some wells, with a mean of 78 mg/L (n = 43). In addition, the spot-wise elevated values reported in certain other locations, such as the Kalpitiya and Jaffna peninsulas, may also be associated with the leaching of contaminants from pit latrines and waste dumps.

Water hardness

Water hardness (Ca^{2+} and Mg^{2+}) is one of the important parameters that have a strong correlation with the geogenic environment. The majority of the groundwater in the dry zone of Sri Lanka belongs to the moderately hard and hard categories (Figure 2 and 3). Health implications related to the water hardness in Sri Lanka are limited except for the incidences of kidney stones in discrete locations in Sri Lanka (Abeywickarama *et al*., 2016). However, the hardness of water and the dominance of its constituents have been suggested as a possible cause of CKDu. In the dry zone of Sri Lanka, specifically in identified CKDu hot spots, the reported hardness was comparatively higher than the non-CKDu areas (Balasooriya *et al*., 2020; Liyanage *et al*., 2022) (Table 2). The high fluoride content associated with high hardness is a significant hydrogeochemical feature in all CKDu regions of Sri Lanka. Other divalent cations such as Sr^{2+} and Ba^{2+} were also found in higher concentrations in groundwater from the dry zone regions.

Compared to Na^+ activity, Ca^{2+} activity is more prominent in CKDu regions. It has been suggested that lower Na^+/Ca^{2+} ratios in groundwater with elevated fluoride levels could exert negative effects on kidney cells (Chandrajith *et al*., 2011a). A recent study revealed a possible influence of Mg^{2+} on CKDu progression (Liyanage *et al*., 2022). It was suggested that the presence of excess fluoride in hard water with increased Mg^{2+} ions possibly triggers the progression of kidney damage. With the availability of constituent ions, the pairing of fluoride and Mg^{2+} in water may form the $(MgF)^+$ complex, which is nephrotoxic, as indicated by considering the Gibbs free energy calculations for ion pair formation and consideration of Hofmeister-type action of anions (Dharma-wardana. 2018). The formed $(MgF)^+$ complex may disrupt the structure and functionality of proteins by breaking apart and being retained on protein surfaces (Dharma-wardana *et al*., 2015). As such, the prolonged consumption of Mg^{2+} rich hard water with fluoride may synergistically influence kidney damage and/or disease progression. Histopathological investigations on the kidney tissues of laboratory mice, treated with high fluoride water for a long period have revealed acute tubular injury (Wasana *et al*., 2017; Perera *et al*., 2021). Even though the initial damage for CKDu may be caused by some other factors, drinking water with excess fluoride and Mg-hardness could enhance the damage further leading to end-stage renal failure. Although higher hardness is evident in sedimentary terrain, low fluoride content does not lead to chronic renal failures.

Table 2: Concentrations of major constituents of groundwater from identified CKDu hotspot regions

Identified CKDu hotspot region		Parameter							Reference
		HD (mg/L)	F^- (mg/L)	Na (mg/L)	Ca (mg/L)	Mg (mg/L)	As (μg/L)	Cd (μg/L)	
Girandurukotte (n = 180)									
	Min	5.64	0.01	0.08	2.07	0.05	< 0.02	< 0.01	Balasooriya *et al*., 2020
	Max	516	5.20	157	394	76.2	2.54	43.0	Wickramarathna *et al*., 2017
	Mean	135	0.74	24.7	31.8	11.2	0.24	0.94	Present study
	Median	107	0.54	18.5	19.5	7.17	0.16	0.02	
Wilgamuwa (n = 344)									
	Min	8.00	0.01	1.31	3.70	0.26	0.02	< 0.01	Wickramarathna *et al*., 2017
	Max	830	4.40	495	1451	472	4.80	11.38	Present study
	Mean	158	0.71	35.3	94.6	19.6	0.47	0.20	
	Median	124	0.60	24.4	31.6	13.5	0.28	0.03	
Nikawewa (n = 11)									
	Min	236	0.16	14.6	13.8	3.06	0.05	< 0.01	Wickramarathna *et al*., 2017
	Max	628	3.44	157	152	74.3	0.30	0.44	
	Mean	397	1.03	64.5	71.1	35.1	0.15	0.11	
	Median	426	0.79	43.9	36.4	35.3	0.15	0.08	
Moneragala (n = 30)									
	Min	16.6	0.28	4.6	12.3	4.23	0.06	< 0.01	Liyanage *et al*., 2022
	Max	162	6.80	204	131	44.2	0.62	0.12	
	Mean	82.4	2.02	35.0	57.7	24.8	0.24	0.03	
	Median	81.8	1.70	22.7	53.1	27.1	0.17	0.03	
Medawachchiya / Medirigiriya (n = 97)									
	Min	NA	0.08	0.75	3.76	0.15	< 1.0	< 0.14	Levine *et al*., 2016
	Max	NA	2.05	156	161	58.4	1.55	0.17	
	Mean	NA	0.60	39.8	55.4	20.5	NA	NA	
	Median	NA	NA	NA	NA	NA	NA	NA	
Padaviya (n = 34)									
	Min	70	0.02	2.09	2.35	2.08	NA	NA	Chandrajith *et al*., 2011
	Max	816	1.33	188	113	41.6	NA	NA	
	Mean	443	0.62	58.3	35.0	20.1	NA	NA	
	Median	NA	NA	NA	NA	NA	NA	NA	

HD: total hardness NA: not available

In addition to that, an inverse relationship between mortality with cardiovascular disease and water hardness has been suggested in earlier studies. A significant risk level of cardiovascular disease has been observed among the population who consume soft water (Knezović *et al*., 2014). Particularly, increased Mg levels in drinking water are considered cardio-protective and may reduce the risk of mortality through cardiovascular disease. As such, water with higher Mg levels may be beneficial to populations that have insufficient Mg intake (Monarca *et al*., 2006; Leurs *et al*., 2010).

Arsenic

Arsenic contamination in groundwater around the world, together with serious associated health problems, has been known for decades and nearly 108 countries are now known to be affected (Shaji *et al*., 2021). The inorganic form of arsenic is extremely toxic and can cause both acute and chronic effects. Skin lesions and pigmentation, gangrene and malignant neoplasm, and hard patches on the palms and soles of the feet are the common symptoms of As poisoning (Smedley & Kinniburg, 2002). Even at lower exposure levels, like 5 μg/L via drinking water, it can cause skin manifestations, while chronic exposure can affect kidneys, liver, and lungs (Yoshida *et al*., 2004).

Elemental arsenic occurs in association with sulphide minerals, iron oxides, and clays (Bhattacharyya *et al*., 2003). Leaching from As-bearing minerals in aquifer material is the main source of inorganic As in groundwater and its mobility is governed by geological, climatological, and geochemical characteristics such as pH and redox conditions. In the Sri Lankan context, arsenic content in groundwater has gained wider attention, as it has been suggested as a possible causative factor for the emergence of CKDu (Jayasumana *et al*., 2014). However, many previous studies have reported low availability of arsenic in groundwater in the metamorphic terrain (Wickramarathna *et al*., 2017; Nanayakkara *et al*., 2019; Liyanage *et al*., 2022). This is further confirmed by the available water quality data, in which none of the samples from metamorphic terrain, particularly from the CKDu areas, exceeded the maximum permissible limit for As. A study on keratinized matrices from patients with chronic kidney diseases of uncertain aetiology (CKDu), has also revealed that CKDu patients were not exposed to toxic levels of As (Diyabalanage *et al*., 2017). Therefore, it is quite unlikely that As is contributing to the onset of CKDu or the disease progression. However, in contrast to metamorphic terrain, higher As levels have been reported in the sedimentary terrain, particularly in Mannar and Kalpitiya regions. It has been revealed that As occurs as coatings in mineral grains in silty clay and sandy layers (Acharyya & Shah, 2007; Amarathunga *et al*., 2019). The As adsorbed to Fe-oxyanions in aquifer material may get mobilized with the pH at a near-neutral level and facilitated by reductive dissolution processes (Amarathunga *et al*., 2019). No data on associated health effects in these regions are yet available; hence further investigations are needed.

CONCLUSIONS

Identifying and quantifying the influence of the geogenic environment on human health is quite challenging. The synergistic or antagonistic activity of elements, and long-term exposure to them, may pose a risk of toxicity. The hydrogeochemical data obtained from the dry zone of Sri Lanka supports improved understanding of the possible geo-environmental influences over the prevailing health issues and draws attention to the critical state faced by the dry zone community. Higher contents of dissolved solids were observed and identified as a typical feature of the dry zone groundwater. Among many others, excess fluoride, hardness, and arsenic were identified as alarming issues. Considerable numbers of water sources were found to be rich in fluoride, mainly due to the underlying geology and the hydrodynamic nature of the aquifer system. Hard water with excess fluoride has been identified as a significant feature in CKDu affected regions, hence the prolonged consumption of such water could affect kidneys. The groundwater in certain sedimentary terrains has been identified as highly vulnerable to nitrate pollution. Although the toxic trace elements such as As, Cd, and Pb were mostly found below the WHO permissible range, elevated arsenic levels were found in the sedimentary terrain. Providing safe drinking water to the community is one of the important aspects defined under the sustainable development goals. However, the current state of the groundwater quality in the dry zone is quite alarming and assessment of and monitoring its chemical composition and pollution levels are essential requirements to provide safe water to the community.

Availability of data and materials

The dataset used in this paper is available with the corresponding author upon a reasonable request.

Competing interests

The authors declare that they have no competing interests.

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